(A) - (4) JP 2002-84992 A

PATENT ABSTRACTS OF JAPAN

(11)Publication number : 2002-084992
(43)Date of publication of application : 26.03.2002

(51)Int.Cl. A23L 1/076
A23L 1/30
A61K 7/00

(21)Application number : 2000-282859
(22)Date of filing : 19.09.2000

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(54) METHOD FOR PRODUCING LIQUID PROPOLIS FOOD COMPOSITION AND SOLID PROPOLIS RAW PROCESSED PRODUCT

(57)Abstract:
PROBLEM TO BE SOLVED: To provide a method for producing an easily eatable alcoholic propolis food composition with water affinity in high extraction efficiency and a solid propolis raw processed product suitable for various uses.
SOLUTION: This method for producing an alcohol solution-type propolis food composition with water affinity comprises such processes that a micelle solution containing a water-based medium and a polyol fatty acid ester-based emulsifier is contacted with a propolis mass and the resultant product is subjected to an extract treatment with ethylalcohol followed by subjected to solid-liquid separation. The other objective solid propolis technical processed product is produced by adsorbing the composition.

LEGAL STATUS

[Date of request for examination]

[Date of sending the examiner's decision of rejection]

[Kind of final disposal of application other than the examiner's decision of rejection or application converted registration]

[Date of final disposal for application]

[Patent number]

[Date of registration]

[Number of appeal against examiner's decision of rejection]

[Date of requesting appeal against examiner's decision of rejection]

[Date of extinction of right]

JAPANESE [JP,2002-084992,A]

CLAIMS DETAILED DESCRIPTION TECHNICAL FIELD PRIOR ART EFFECT OF THE INVENTION TECHNICAL PROBLEM MEANS EXAMPLE

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CLAIMS

[Claim(s)]
[Claim 1] The manufacture approach of the solid-like propolis original object workpiece which comes to adsorb the water compatibility alcoholic solution mold propolis food constituent and it which perform extract processing by ethyl alcohol and are characterized by subsequently carrying out solid liquid separation after contacting the micellar solution containing a drainage system medium, and polyol and a fatty-acid-ester system emulsifier to a propolis original lump.
[Claim 2] The approach according to claim 1 of being the mixture of the water-soluble compound in which a drainage system medium can carry out hydrogen bond to a water independent or water, and water in multiplex.
[Claim 3] The approach according to claim 1 or 2 by which solid-like propolis original object workpiece is used as an ingredient of the charge for the skins of makeup.
[Claim 4] The approach according to claim 1 or 2 by which solid-like propolis original object workpiece is used as a plant growth regulator.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]
[0001]
[Field of the Invention] This invention relates to the manufacture approach of a solution mold propolis food constituent and solid-like propolis original object workpiece. In more detail, this invention is a thing about an advantageous approach which adsorbs the above-mentioned propolis food constituent and also manufactures useful solid-like propolis original object workpiece for various applications industrially at the same time it manufactures the propolis food constituent of an alcoholic solution mold which has the water compatibility which is easy to eat with sufficient extraction efficiency.
[0002]
[Description of the Prior Art] The propolis known as a natural antimicrobial agent for many years is the quality of a solid of the shape of resin from which the honeybee mixed a honeybee's own secrete, a bee wax, etc. in the matter list of the specific part of a tree, the gums extracted mainly from the sprout, or a bud and a bark, sap, and a vegetable pigment system, and was made by aggregates, such as perfumed oil, at it. When eating this, if it remains as it is, since it is hard and unsuitable, by the solution extract which usually uses ethyl alcohol, a liquefaction carbon dioxide, water, liquefied polysaccharide, etc., it considered as solution-like food and has been offered by making it the gestalt which can be further diluted with a medium.
[0003] In these extract approaches, with the liquefaction carbon-dioxide extraction method, there was a problem that a component sampling volume was low, the water extraction method and the liquefied polysaccharide extraction method made the component solidified after the extract remelt in other media, and the process changed into the gestalt which is easy to eat was needed, and they had the problem that actuation was complicated. From such a situation, there are many component sampling volumes and, moreover, former most extraction methods by ethyl alcohol with easy actuation have been performed.
[0004] by the way, in case propolis food eats it It is desirable to have the property dissolved or distributed in water, since it dilutes with water and puts into inner mouth. In the semantics Since the propolis food obtained by the ethyl alcohol extraction method was nonaqueous solubility, it remained the component's having deposited and adhering to a container, when diluted with water, or the situations which are not desirable, such as shifting to neither the stomach nor intestines, were smoothly invited from opening and an esophagus, and the amelioration was desired.
[0005] Then, in order that this invention persons may aim at improvement in compatibility with the water of ethyl alcohol extract propolis food By contacting the micellar solution containing a drainage system medium, and making it mix ethyl alcohol extract propolis food with polyol and a fatty-acid-ester system emulsifier previously How (JP,4-66544,B) to manufacture the propolis food constituent which reforms the front face of an extract component and presents water-dispersion, How to add a saponin in ethyl alcohol extract propolis food (JP,6-197734,A), Ethyl alcohol extract propolis food was contacted in the grain protein partial decomposition product solution, and the approach (JP,9-75018,A) of giving water-dispersion etc. was found out.
[0006] On the other hand, in order to manufacture a propolis food constituent with good compatibility with water from the start, in another invention in above-mentioned JP,4-66544,B, this invention persons showed the manufacture approach of contacting the propolis original lump itself to the drainage system medium micell of polyol and a fatty-acid-ester system emulsifier, and succeeded in producing the propolis food constituent of a water solubilization mold at one process. However, although this approach might satisfy the compatibility with water extremely, it is the point of a component sampling volume and had the problem that it was inferior compared with ethyl alcohol extract propolis food. Moreover, in the manufacture approach of the propolis food constituent by the conventional simple ethyl alcohol extract, generally insoluble propolis residue (extract residue) is disposed of as industrial waste, and development of the deployment was desired.
[0007]
[Problem(s) to be Solved by the Invention] This invention is the basis of such a situation and aims at offering the manufacture approach of useful insoluble propolis residue (extract residue) for various applications as the propolis food constituent and the functional product of the alcoholic solution mold which can aim at both amelioration of the affinity of improvement in extraction efficiency, and water to coincidence according to the consistent process from a propolis original lump.
[0008]
[Means for Solving the Problem] In order that this invention persons may attain said purpose, as a result of repeating research wholeheartedly, first After making a drainage system medium and the micellar solution containing polyol and a fatty-acid-ester system emulsifier contact, permeating and making a propolis original lump fully adsorb, by performing extract processing with ethyl alcohol The alcoholic solution from which the osmosis operation into the propolis original lump of ethyl alcohol and the extract of a component, and the solvent action improved, and extract residue was moreover removed according to solid liquid separation It becomes the alcoholic solution mold propolis food constituent which has water compatibility more stable than the thing which made the emulsifier component contain by adding after mixing to the conventional ethyl alcohol extract propolis food constituent. And it found out that the above-mentioned extract residue adsorbed this constituent, and served as useful solid-like propolis original object workpiece as a functional product at various applications. This invention is completed based on these knowledge.
[0009] That is, this invention offers the manufacture approach of the solid-like propolis original object workpiece which comes to adsorb the water compatibility alcoholic solution mold propolis food constituent and it which perform extract processing by ethyl alcohol and are characterize by subsequently carry out solid liquid separation, after contact the micellar solution containing a drainage system medium, and polyol and a fatty acid ester system emulsifier to a propolis original lump.
[0010]

[Embodiment of the Invention] It is not restricted especially as a propolis original lump of the raw material in the approach of this invention, but you may be the thing of what kind of the origin, for example, the product from Brazil, the product from the U.S., the product from Germany, the product from China, the product from Australia, etc. can use all.
[0011] In the approach of this invention, the micellar solution containing a drainage system medium, and polyol and a fatty-acid-ester system emulsifier is first contacted to the above-mentioned propolis original lump. as the drainage system medium in the above-mentioned micellar solution -- water -- it may be independent and you may be the mixture of the water-soluble compound which can carry out hydrogen bond to water and water in multiplex. Here, as an example of the water-soluble compound which can carry out hydrogen bond to water in multiplex, propylene glycol, a glycerol, D-xylose, D-sorbitol, a citric acid, a malic acid, a succinic acid, a fumaric acid, a tartaric acid, a gluconic acid, a glucono delta lactone, a glycine, etc. can be mentioned. These may use one sort independently and may use it combining two or more sorts.
[0012] On the other hand, as polyol and a fatty-acid-ester system emulsifier For example, (i) Glycerol = [Mono-laurate glycerol = monopalmitate,] Glycerol = Monostearate, glycerol = monooleate, a glycerol = MONORINO rate, Glycerol = The fatty acid ester of glycerols, such as mono-triricinolate, (ii) -- diglycerol = -- mono-laurate and diglycerol = monopalmitate -- Diglycerol = Monostearate, diglycerol = monooleate, Diglycerol = A MONORINO rate, diglycerol = mono-triricinolate, Tetra-glycerol = Mono-laurate, a tetra-glycerol = JIRAU rate, Tetra-glycerol = Monopalmitate, tetra-glycerol = dipalmitate, Tetra-glycerol = Monostearate, tetra-glycerol = distearate, Tetra-glycerol = Monooleate, tetra-glycerol = dioleate, Tetra-glycerol = A MONORINO rate, a tetra-glycerol = JIRINO rate, Tetra-glycerol = Mono-triricinolate, a tetra-glycerol = JIRISHINO rate, Tetra-glycerol = Mono-behenate, tetra-glycerol = dibehenate, PENTA glycerol = Mono-laurate, a PENTA glycerol = JIRAU rate, PENTA glycerol = Monopalmitate, PENTA glycerol = dipalmitate, PENTA glycerol = Monostearate, PENTA glycerol = distearate, PENTA glycerol = Monooleate, PENTA glycerol = dioleate, PENTA glycerol = A MONORINO rate, a PENTA glycerol = JIRINO rate, PENTA glycerol = Mono-triricinolate, a PENTA glycerol = JIRISHINO rate, PENTA glycerol = Mono-behenate, PENTA glycerol = dibehenate, Deca glycerol = Mono-laurate, a deca glycerol = JIRAU rate, Deca glycerol = Trilaurate, deca glycerol = monopalmitate, Deca glycerol = Dipalmitate, deca glycerol = tripalmitate, Deca glycerol = Monostearate, deca glycerol = distearate, Deca glycerol = Tristearate, decaglycerin monooleate, Deca glycerol = Dioleate, deca glycerol = trio REETO, Deca glycerol = A MONORINO rate, a deca glycerol = JIRINO rate, Deca glycerol = A TORIRINO rate, deca glycerol = mono-triricinolate, Deca glycerol = A JIRISHINO rate, a deca glycerol = TORIRISHINO rate, Deca glycerol = Mono-behenate, deca glycerol = dibehenate, Deca glycerol = Tribehenate, deca glycerol = monoisostearate, Deca glycerol = Sesqui-isostearate, deca glycerol = diisostearate, Deca glycerol = TORIISO stearate, deca glycerol = monochrome (12-hydroxy) stearate, Deca glycerol = the fatty acid ester of polyglycerin, such as JI (12-hydroxy) stearate and deca glycerol = Tori (12-hydroxy) stearate, [0013] (iii) Propylene glycol = [Mono-laurate, propylene glycol = monopalmitate,] Propylene glycol = Monostearate, propylene glycol = monooleate, Propylene glycol = A MONORINO rate, propylene glycol = mono-triricinolate, Propylene glycol = The fatty acid ester of propylene glycols, such as monoisostearate and propylene glycol = monochrome (12-hydroxy) stearate, (iv) Sorbitan monolaurate, a sorbitan = JIRAU rate, Sorbitan monopalmitate, sorbitan = dipalmitate, sorbitan = monostearate, Sorbitan = Distearate, sorbitan monooleate, sorbitan = dioleate, Sorbitan = A MONORINO rate, a sorbitan = JIRINO rate, sorbitan = mono-triricinolate, Sorbitan = A JIRISHINO rate, sorbitan = mono-behenate, sorbitan dibehenate, The fatty acid ester of sorbitan, such as sorbitan monoisostearate, sorbitan = diisostearate, sorbitan = monochrome (12-hydroxy) stearate, and sorbitan = JI (12-hydroxy) stearate, [0014] (v) Cane sugar = [Mono-laurate, a cane-sugar = JIRAU rate, cane-sugar = monopalmitate,] Cane sugar = Dipalmitate, cane-sugar = monostearate, cane-sugar = distearate, Cane sugar = Monooleate, cane-sugar = dioleate, a cane-sugar = MONORINO rate, Cane sugar = A JIRINO rate, cane-sugar = mono-triricinolate, a cane-sugar = JIRISHINO rate, Cane sugar = lecithin, such as fatty acid ester of cane sugars, such as monoisostearate, cane-sugar = diisostearate, cane-sugar = monochrome (12-hydroxy) stearate, and cane-sugar = JI (12-hydroxy) steer **-TO, and (vi) soybean-oil lecithin, etc. is mentioned. These may use one sort independently and may use it combining two or more sorts.
[0015] In this invention, when using the mixture of the water-soluble compound which can carry out hydrogen bond to water and water in multiplex as said drainage system medium, as for the content of the water in this mixture, it is desirable that it is 10 % of the weight or more. It is difficult for the content of water to make polyol and a fatty-acid-ester system emulsifier a micell dissolution condition at less than 10 % of the weight, a desired micellar solution is hard to be obtained, and there is a possibility that quick and sufficient adsorption on a propolis original lump front face may not be performed. Consequently, in the extract processing by the ethyl alcohol in degree process, since amelioration of the affinity of the improvement in extraction efficiency and water which a micell catalyst device is not fully demonstrated by the REBINDA effectiveness list, but may be satisfied cannot be achieved, it is not desirable.
[0016] Moreover, in preparation of a micellar solution, it is good 0.1 - 20 weight section and to dissolve preferably per said drainage system medium 100 weight section, and polyol and a fatty-acid-ester system emulsifier at a rate of 1 - 5 weight section. Since there will be a possibility that a micell gestalt may change and it will become inadequate [all] sticking to a propolis original lump front face if it is hard to carry out the micell dissolution of this emulsifier under in the 0.1 weight section and the amount of dissolutions of polyol and a fatty-acid-ester system emulsifier exceeds 20 weight sections, it is not desirable.
[0017] As for this micellar solution, in this invention, it is advantageous to make it contact at a rate of 15 weight sections at least to the propolis original lump 100 weight section. Infiltration on a propolis original lump front face and adsorption are fully hard to perform the amount of a micellar solution under in 15 weight sections. This infiltration and adsorption actuation are usually performed using Brabender, a kneader, etc., and 30-100 degrees C is usually preferably selected from the point of workability in 50-70 degrees C as temperature in that case.
[0018] Next, after doing in this way and contacting said micellar solution to a propolis original lump, extract processing by ethyl alcohol is performed. per propolis original lump 100 weight section from the point which extracts smoothly and raises a quantitative formula in this extract processing, and ethyl alcohol -- usually -- the 100 - 1000 weight section -- the 200 - 500 weight section comes out comparatively preferably, and it uses.
[0019] In this invention, since a micell catalyst device works automatically also in the state of standing, an extract is also made quickly, but if the whole system is heated or it shakes or mixes [stirring], the REBINDA effectiveness will be demonstrated better, consequently extract processing will be accelerated further.
[0020] In this invention, after doing in this way and performing extract processing, solid liquid separation is usually performed to the bottom of ordinary temperature, and it separates into an alcoholic solution and insoluble propolis residue (extract residue). There is especially no limit as the approach of solid liquid separation, and filtration, a centrifuge method, etc. using a well-known approach, for example, the decantation method, a filter paper, a filter cloth, a wire gauze, etc. can be used conventionally. It is offered as solid-like propolis original object workpiece with which the alcoholic above-mentioned solution adsorbs as a water

compatibility alcoholic solution mold propolis food constituent, and extract residue comes to adsorb this constituent.
[0021] This water compatibility alcoholic solution mold propolis food constituent makes the amount of the ethyl alcohol in it fluctuate on the occasion of commercial production, and can be adjusted to suitable concentration and viscosity. Thus, water compatibility is good and the alcoholic solution mold propolis food constituent which is easy to eat is obtained with sufficient extraction efficiency.
[0022] Moreover, solid-like propolis original object workpiece is variously useful as a functional product as an ingredient, a plant growth regulator, etc. of the application of makeup, for example, the charge for the skins. In addition, generally in the manufacture approach of the propolis food constituent by the conventional simple ethyl alcohol extract, extract residue is disposed of as industrial waste.
[0023]
[Example] Next, although an example explains this invention to a detail further, this invention is not limited at all by these examples.
[0024] After teaching the propolis original lump 100 weight section from Brazil to example 1 Brabender, the micellar solution which consists of the water 100 weight section, the glycerol 10 weight section, and the tetra-glycerol = mono-laurate 0.4 weight section is added, under ordinary temperature, by 1000rpm, stirring was performed for 15 minutes and mutual contact was carried out. Subsequently, this contact processing object whole quantity was moved to the Erlenmeyer flask with ground-in stopper, the ethyl alcohol 300 weight section adjusted to 20 degrees C at this was added, and standing was carried out for two weeks in 20-degree-C thermostatic chamber. Then, while processing for 30 minutes by 1000rpm under ordinary temperature, extracting the upper supernatant and obtaining the water compatibility alcoholic solution mold propolis food constituent of this invention with a centrifugal separator, the solid-like residue in the condition of having adsorbed it was extracted from the lower layer.
[0025] The propolis original lump 100 weight section from Brazil was put into the example of comparison 1 Erlenmeyer flask with ground-in stopper, the ethyl alcohol 300 weight section subsequently to 20 degrees C adjusted was added, and standing was carried out for two weeks in 20-degree-C thermostatic chamber. Then, while processing for 30 minutes by 1000rpm, extracting the supernatant and obtaining an alcoholic solution mold propolis food constituent under ordinary temperature with a centrifugal separator, lower layer solid-like residue was extracted.
[0026] About the propolis food constituent obtained in the example 1 and the example 1 of a comparison, the total amount of flavonoid was measured and the propolis quantitative formula was computed. Moreover, it put and water compatibility was checked, after adding to underwater [which was adjusted to 20 degrees C / 100g] and mixing 5g of each propolis food constituent for 1 minute. A result is shown in Table 1.
[0027]
[Table 1]

表1

試　料	プロポリス成分含有量（重量%）	水親和性		
		混合直後	1日静置後	5日静置後
実施例1のﾌﾟﾛﾎﾟﾘｽ食品組成物	24.5	均一乳化状態	均一乳化状態	均一乳化状態
比較例1のﾌﾟﾛﾎﾟﾘｽ食品組成物	19.7	凝集分離（液白濁）	凝集分離（液半透明）	凝集分離（液透明）

[0028] As for the propolis food constituent of an example 1, it turns out that a sampling volume also has water compatibility a good top, and tends to eat it so that clearly from Table 1.
[0029] The micellar solution which consists of the sorbitan monolaurate 0.3 weight section, the cane-sugar = monooleate 0.2 weight section, and the water 50 weight section is added, after teaching the propolis original lump 100 weight section from Australia to the 4 opening flask equipped with measuring tubing linked to example 2 agitator, a thermometer, a gas inhalant canal, and a capacitor, at 50-60 degrees C, stirring is performed for 2 hours, it contacted and it was mixed. Subsequently, the ethyl alcohol 800 weight section is added to this, and stirring is performed at 50-60 degrees C for 1 hour, and a shaking and after making it mix, passed nitrogen gas, the ethyl alcohol of the 500 weight sections was made to distill off, stirring was stopped, and it was made to cool to ordinary temperature. Then, while filtering under ordinary temperature using the No. 2 filter paper, extracting the filtrate and obtaining the water compatibility alcoholic solution mold propolis food constituent of this invention, the solid-like residue in the condition of having adsorbed it was extracted from the filter paper.
[0030] The propolis original lump 100 weight section from Australia is taught, the ethyl alcohol 800 weight section is added further, stirring is performed at 50-60 degrees C for 1 hour, a shaking and after making it mix, nitrogen gas was passed in the 4 opening flask equipped with measuring tubing linked to example of comparison 2 agitator, a thermometer, a gas inhalant canal, and a capacitor, and it was made to distill off the ethyl alcohol of the 500 weight sections. Subsequently, add the sorbitan monolaurate 0.3 weight section and the cane-sugar = monooleate 0.2 weight section to this, it was made to dissolve in it in 50-60 degrees C, stirring was stopped, and it was made to cool to ordinary temperature. Then, while filtering under ordinary temperature using the No. 2 filter paper, extracting the filtrate and obtaining an alcoholic solution mold propolis food constituent, solid-like residue was extracted from the filter paper.
[0031] While measuring the propolis quantitative formula like the example 1, respectively about the propolis food constituent obtained in the example 2 and the example 2 of a comparison, the water compatibility trial was performed. A result is shown in Table 2.
[0032]
[Table 2]

表2

試料	プロポリス成分含有量（重量%）	水親和性		
		混合直後	1日静置後	5日静置後
実施例2のﾌﾟﾛﾎﾟﾘｽ食品組成物	36.5	均一乳化状態	均一乳化状態	均一乳化状態
比較例2のﾌﾟﾛﾎﾟﾘｽ食品組成物	28.9	均一分散状態	上層に分離液が形成	下層に沈殿が形成

[0033] In the example 2 which is carrying out extract processing of the solution which comes to carry out the micell dissolution of the emulsifier component into a drainage system medium with ethyl alcohol since a propolis original lump is contacted beforehand , an emulsifier component is understood that the top where the extraction efficiency of a propolis component is high and water compatibility are good compared with the example 2 of a comparison which is carrying out extract processing by the system which carries out the simple dissolution in ethyl alcohol so that clearly from Table 2 .

[0034] The propolis original lump of a class and an amount which shows in Table 3 and 4 according to the approach of three to example 10 example 1, or an example 2. By performing extract processing by ethyl alcohol after contact processing on the conditions shown in Table 3 and 4 using a drainage system medium, and polyol and a fatty-acid-ester system emulsifier, and subsequently performing after treatment While obtaining the water compatibility alcoholic solution mold propolis food constituent, the solid-like residue in the condition of having adsorbed it was extracted.

[0035]

[Table 3]

表3

実施例	プロポリス原塊	媒体	ポリオール・脂肪酸エステル系乳化剤	エチルアルコール	製造方法
3	ブラジル産プロポリス原塊 100重量部	水 15重量部	ｸﾞﾘｾﾘﾝ=ﾓﾉﾗｳﾚｰﾄ 0.05重量部 ﾍﾟﾝﾀｸﾞﾘｾﾘﾝ=ﾓﾉｽﾃｱﾚｰﾄ 0.1重量部	500重量部投入し、常温で48時間静置、200重量部を系外に除去した。	実施例1の製造方法に準ずる
4	米国産プロポリス原塊 100重量部	水 15.5重量部 D-ｿﾙﾋﾞｯﾄ 3重量部 ｹﾞﾙｺﾉﾃﾞﾙﾗﾗｸﾄﾝ 1重量部 ﾌﾟﾛﾋﾟﾚﾝｸﾞﾘｺｰﾙ 0.5重量部	ﾌﾟﾛﾋﾟﾚﾝｸﾞﾘｺｰﾙ ・ﾓﾉﾗｳﾚｰﾄ 0.01重量部 ｼﾞｸﾞﾘｾﾘﾝ=ﾓﾉﾘｼﾉﾚｰﾄ 0.01重量部	300重量部投入し、常温で72時間静置した。	同上
5	ドイツ産プロポリス原塊 100重量部	水 3重量部 ｸｴﾝ酸 1重量部 ｸﾞﾘｾﾘﾝ 26重量部	ｼｮ糖=ﾓﾉﾊﾟﾙﾐﾃｰﾄ 2重量部 ﾃﾞｶｸﾞﾘｾﾘﾝ=ﾓﾉﾘﾉﾚｰﾄ 4重量部	300重量部投入し、40℃で15時間静置した。	同上
6	中国産プロポリス原塊 100重量部	水 15重量部 D-ｷｼﾛｰｽ 4重量部 ｸﾞﾘｼﾝ 1重量部	ｿﾙﾋﾞﾀﾝ=ﾓﾉｵﾚｴｰﾄ 0.04重量部 ﾃﾄﾗｸﾞﾘｾﾘﾝ=ﾓﾉ12-ﾋﾄﾞﾛｷｼｽﾃｱﾚｰﾄ 0.04重量部	200重量部投入し、50℃で24時間静置した後、さらに、常温で100重量部投入した。	同上

[0036]

[Table 4]

表4

実施例	プロポリス原塊	媒体	ポリオール・脂肪酸エステル系乳化剤	エチルアルコール	製造方法
7	米国産プロポリス原塊 100重量部	水 23重量部 リンゴ酸 2重量部	ペンタグリセリン・ジラウレート 0.4重量部 ショ糖・モノステアレート 0.4重量部	300重量部投入し、60℃で2時間、撹拌、混合させた。	実施例2の製造方法に準ずる
8	ドイツ産プロポリス原塊 100重量部	水 30重量部 クエン酸 2重量部 グルコン酸 1重量部 グリセリン 7重量部	ショ糖=モノオレート 3重量部 ペンタグリセリン・モノイソステアレート 4重量部 大豆油レシチン 1重量部	800重量部投入し、常温で24時間撹拌・混合させた後、500重量部を系外に除去した。	同上
9	中国産プロポリス原塊 100重量部	水 5重量部 D-キシロース 5重量部 グリセリン 40重量部	グリセリン=モノリシノレート 0.2重量部 テトラグリセリン=ジラウレート 0.2重量部	300重量部投入し、常温で24時間、撹拌・混合させた。	同上
10	オーストラリア産プロポリス原塊 100重量部	水 35重量部 D-ソルビット 12重量部 酒石酸 3重量部	ソルビタン=モノラウレート 0.03重量部 ショ糖=ジラウレート 0.02重量部	300重量部投入し、50℃で3時間、撹拌・混合させた。	同上

[0037] While measuring the propolis quantitative formula like the example 1 about the propolis food constituent obtained in the examples 3-10, respectively, the water compatibility trial was performed. A result is shown in Table 5.

[0038]

[Table 5]

表5

試料	プロポリス成分含有量（重量%）	水親和性		
		混合直後	1日静置後	5日静置後
実施例3のプロポリス食品組成物	17.1	均一乳化状態	均一乳化状態	均一乳化状態
実施例4のプロポリス食品組成物	16.3	同上	同上	同上
実施例5のプロポリス食品組成物	19.2	同上	同上	同上
実施例6のプロポリス食品組成物	16.1	同上	同上	同上
実施例7のプロポリス食品組成物	18.8	同上	同上	同上
実施例8のプロポリス食品組成物	18.9	同上	同上	同上
実施例9のプロポリス食品組成物	17.5	同上	同上	同上
実施例10のプロポリス食品組成物	16.2	同上	同上	同上

[0039] Each propolis food constituent of examples 3-10 has the good extraction efficiency of a propolis component, and the water compatibility made into the purpose also comes out enough, and understands a certain thing for it so that clearly from

Table 5.
[0040] The water 12 weight section, the glycerol 64.7 weight section, the stearin acid 8 weight section, and the sodium-hydroxide 0.3 weight section were taught to the beaker which attached application 1 agitator and the thermometer, and it mixed, heating gradually, and the milky lotion was prepared at 80 degrees C. Subsequently, after lowering an internal temperature to 50 degrees C, the dirt dropping cream of a dispersion mold was produced by supplying the solid-like residue 15 weight section obtained in the example 1, and mixing homogeneity for 30 minutes by 1000rpm.
[0041] In comparison application 1 application 1, the dirt dropping cream of a dispersion mold was produced like the application 1 except having used the solid-like residue obtained in the example 1 of a comparison instead of the solid-like residue obtained in the example 1.
[0042] Standing was carried out to the thermostat which moved 10g of dirt dropping creams of an application 1 and the comparison application 1 at a time to the glass cylinder, respectively, and adjusted them to 25 degrees C for one month, and the stability of dispersion was observed.
[0043] I had the feeling after the dirt dropping effectiveness after having divided into ten persons at a time 20 persons who chose at random about each dirt dropping cream, having the arm and the hand use them after activity termination and having them subsequently wiped off with a cloth towel, a touch rate, and use told on the other hand out of the 18-60-year-old man who is doing the field work about the thing immediately after manufacture in the car garage for working 8 hours for one day. These results are shown in Table 6.
[0044]
[Table 6]

表6

試　料	ﾃﾞｨｽﾊﾟｰｼﾞｮﾝ安定性（２５℃、１ヶ月後）	汚れ落とし効果　1)	肌触わり　2)	使用後の感覚　3)
応用例１の汚れ落としクリーム	均一状態	○	○	○
比較応用例１の汚れ落としクリーム	底部に沈殿物形成	△	×	△

[0045] (Note)
1) The inside of the thing **:10 person examiner who eight or more persons' person accepted was good among the valuation-basis O:10 person [of the dirt dropping effectiveness] examiner, Those who admited being good among the thing x:10 person examiner who 5-7 persons accepted was good The inside of a valuation-basis O:10 person [of four or less persons' thing 2 touch rate] examiner, The inside of the thing x:10 person examiner who 5-7 persons' person accepted was good among the thing **:10 person examiner who eight or more persons' person accepted was good, The inside of an evaluation O:10 person [of the feeling after the thing 3 use which six or more persons' person accepted to be a poor touch rate] examiner, Those who conceded carried out gently among the thing **:10 person examiner who eight or more persons' person accepted carried out gently, and the thing x:10 person examiner who 5-7 persons accepted carried out gently are four or less persons' things [0046]. After sprinkling 2kg of solid-like residue obtained in the example 2 on the average to the place to 20cm of surfaces with application 2 above sea level of 300m of Kanto loam layer Hataji 12m2, 50 seedlings of lavender were planted and it was made to grow them for summer three months.
[0047] After sprinkling 2kg of solid-like residue obtained in the example 2 of a comparison on the average to the place to 20cm of surfaces of Kanto loam layer Hataji 12m2 which adjoin the location in comparison application 2 application 2, 50 seedlings of lavender were planted and it was grown for the same stage three months with the application 2. Moreover, 50 seedlings of lavender were planted in Kanto loam layer Hataji 12m2 which adjoin an application 2 and the comparison application 2, and it was made to grow for three months as it is at a coincidence term. Each growth situation is summarized and it is shown in Table 7.
[0048]
[Table 7]

表7

試　料	頭頂の花部までの長さ			茎部の状態　1)
	45cmより高いもの	35～45cmのもの	35cm未満のもの	
無処理地区のラベンダー	０本／５０本	１２本／５０本	３８本／５０本	C
応用例２のラベンダー	２３本／５０本	２６本／５０本	１本／５０本	A
比較応用例２のラベンダー	３本／５０本	２８本／５０本	１９本／５０本	B

[0049] (Note)
1) The thing C to which 30-39 are strongly extended towards the upper part among A:50 valuation bases of the condition of a scapus among B:50 things to which 40 or more are strongly extended towards the upper part : it turns out that the growth situation of the lavender in an application 2 is good so that clearly [what is strongly extended towards the upper part] from 29 or less thing tables 7.
[0050]

[Effect of the Invention] The wrapped-in component is taken out quickly and efficiently and the reattachment is further controlled in the condensation list within a system at the same time according to this invention a micell catalyst device and the REBINDA effectiveness work and it crushes a propolis original lump by carrying out extract processing with ethyl alcohol, since the drainage system micellar solution of polyol and a fatty-acid-ester system emulsifier is made to stick to the front face of the propolis original lump of a start raw material beforehand.

[0051] Moreover, the extract residue separated and obtained in the final process in the micellar solution adsorption propolis original lump list at the time of the middle process in the manufacture approach of this invention also holds penetrating power. Therefore, since the possibility of an activity of the insoluble matter in the propolis original lump which a change of state is not only made to the water compatibility which the extraction efficiency of an alcoholic extract product goes up, and is moreover easy to eat by carrying out this invention, but had become industrial waste in manufacture of the simple alcoholic conventional extract propolis food is also found out, it becomes a big advantage.

[Translation done.]

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TECHNICAL FIELD

[Field of the Invention] This invention relates to the manufacture approach of a solution mold propolis food constituent and solid-like propolis original object workpiece. In more detail, this invention is a thing about an advantageous approach which adsorbs the above-mentioned propolis food constituent and also manufactures useful solid-like propolis original object workpiece for various applications industrially at the same time it manufactures the propolis food constituent of an alcoholic solution mold which has the water compatibility which is easy to eat with sufficient extraction efficiency.

[Translation done.]

PRIOR ART

[Description of the Prior Art] The propolis known as a natural antimicrobial agent for many years is the quality of a solid of the shape of resin from which the honeybee mixed a honeybee's own secrete, a bee wax, etc. in the matter list of the specific part of a tree, the gums extracted mainly from the sprout, or a bud and a bark, sap, and a vegetable pigment system, and was made by aggregates, such as perfumed oil, at it. When eating this, if it remains as it is, since it is hard and unsuitable, by the solution extract which usually uses ethyl alcohol, a liquefaction carbon dioxide, water, liquefied polysaccharide, etc., it considered as solution-like food and has been offered by making it the gestalt which can be further diluted with a medium.
[0003] In these extract approaches, with the liquefaction carbon-dioxide extraction method, there was a problem that a component sampling volume was low, the water extraction method and the liquefied polysaccharide extraction method made the component solidified after the extract remelt in other media, and the process changed into the gestalt which is easy to eat was needed, and they had the problem that actuation was complicated. From such a situation, there are many component sampling volumes and, moreover, former most extraction methods by ethyl alcohol with easy actuation have been performed.
[0004] by the way, in case propolis food eats it It is desirable to have the property dissolved or distributed in water, since it dilutes with water and puts into inner mouth. In the semantics Since the propolis food obtained by the ethyl alcohol extraction method was nonaqueous solubility, it remained the component's having deposited and adhering to a container, when diluted with water, or the situations which are not desirable, such as shifting to neither the stomach nor intestines, were smoothly invited from opening and an esophagus, and the amelioration was desired.
[0005] Then, in order that this invention persons may aim at improvement in compatibility with the water of ethyl alcohol extract propolis food By contacting the micellar solution containing a drainage system medium, and making it mix ethyl alcohol extract propolis food with polyol and a fatty-acid-ester system emulsifier previously How (JP,4-66544,B) to manufacture the propolis food constituent which reforms the front face of an extract component and presents water-dispersion, How to add a saponin in ethyl alcohol extract propolis food (JP,6-197734,A), Ethyl alcohol extract propolis food was contacted in the grain protein partial decomposition product solution, and the approach (JP,9-75018,A) of giving water-dispersion etc. was found out.
[0006] On the other hand, in order to manufacture a propolis food constituent with good compatibility with water from the start, in another invention in above-mentioned JP,4-66544,B, this invention persons showed the manufacture approach of contacting the propolis original lump itself to the drainage system medium micell of polyol and a fatty-acid-ester system emulsifier, and succeeded in producing the propolis food constituent of a water solubilization mold at one process. However, although this approach might satisfy the compatibility with water extremely, it is the point of a component sampling volume and had the problem that it was inferior compared with ethyl alcohol extract propolis food. Moreover, in the manufacture approach of the propolis food constituent by the conventional simple ethyl alcohol extract, generally insoluble propolis residue (extract residue) is disposed of as industrial waste, and development of the deployment was desired.

[Translation done.]

EFFECT OF THE INVENTION

[Effect of the Invention] The wrapped-in component is taken out quickly and efficiently and the reattachment is further controlled in the condensation list within a system at the same time according to this invention a micell catalyst device and the REBINDA effectiveness work and it crushes a propolis original lump by carrying out extract processing with ethyl alcohol, since the drainage system micellar solution of polyol and a fatty-acid-ester system emulsifier is made to stick to the front face of the propolis original lump of a start raw material beforehand.
[0051] Moreover, the extract residue separated and obtained in the final process in the micellar solution adsorption propolis original lump list at the time of the middle process in the manufacture approach of this invention also holds penetrating power. Therefore, since the possibility of an activity of the insoluble matter in the propolis original lump which a change of state is not only made to the water compatibility which the extraction efficiency of an alcoholic extract product goes up, and is moreover easy to eat by carrying out this invention, but had become industrial waste in manufacture of the simple alcoholic conventional extract propolis food is also found out, it becomes a big advantage.

[Translation done.]

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TECHNICAL PROBLEM

[Problem(s) to be Solved by the Invention] This invention is the basis of such a situation and aims at offering the manufacture approach of useful insoluble propolis residue (extract residue) for various applications as the propolis food constituent and the functional product of the alcoholic solution mold which can aim at both amelioration of the affinity of improvement in extraction efficiency, and water to coincidence according to the consistent process from a propolis original lump.

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MEANS

[Means for Solving the Problem] In order that this invention persons may attain said purpose, as a result of repeating research wholeheartedly, first After making a drainage system medium and the micellar solution containing polyol and a fatty-acid-ester system emulsifier contact, permeating and making a propolis original lump fully adsorb, by performing extract processing with ethyl alcohol The alcoholic solution from which the osmosis operation into the propolis original lump of ethyl alcohol and the extract of a component, and the solvent action improved, and extract residue was moreover removed according to solid liquid separation It becomes the alcoholic solution mold propolis food constituent which has water compatibility more stable than the thing which made the emulsifier component contain by adding after mixing to the conventional ethyl alcohol extract propolis food constituent, And it found out that the above-mentioned extract residue adsorbed this constituent, and served as useful solid-like propolis original object workpiece as a functional product at various applications. This invention is completed based on these knowledge.
[0009] That is, this invention offers the manufacture approach of the solid-like propolis original object workpiece which comes to adsorb the water compatibility alcoholic solution mold propolis food constituent and it which perform extract processing by ethyl alcohol and are characterize by subsequently carry out solid liquid separation, after contact the micellar solution containing a drainage system medium, and polyol and a fatty acid ester system emulsifier to a propolis original lump.
[0010]
[Embodiment of the Invention] It is not restricted especially as a propolis original lump of the raw material in the approach of this invention, but you may be the thing of what kind of the origin, for example, the product from Brazil, the product from the U.S., the product from Germany, the product from China, the product from Australia, etc. can use all.
[0011] In the approach of this invention, the micellar solution containing a drainage system medium, and polyol and a fatty-acid-ester system emulsifier is first contacted to the above-mentioned propolis original lump. as the drainage system medium in the above-mentioned micellar solution — water — it may be independent and you may be the mixture of the water-soluble compound which can carry out hydrogen bond to water and water in multiplex. Here, as an example of the water-soluble compound which can carry out hydrogen bond to water in multiplex, propylene glycol, a glycerol, D-xylose, D-sorbitol, a citric acid, a malic acid, a succinic acid, a fumaric acid, a tartaric acid, a gluconic acid, a glucono delta lactone, a glycine, etc. can be mentioned. These may use one sort independently and may use it combining two or more sorts.
[0012] On the other hand, as polyol and a fatty-acid-ester system emulsifier For example, (i) Glycerol = [Mono-laurate glycerol = monopalmitate,] Glycerol = Monostearate, glycerol = monooleate, a glycerol = MONORINO rate, Glycerol = The fatty acid ester of glycerols, such as mono-triricinolate, (ii) — diglycerol = — mono-laurate and diglycerol = monopalmitate — Diglycerol = Monostearate, diglycerol = monooleate, Diglycerol = A MONORINO rate, diglycerol = mono-triricinolate, Tetra-glycerol = Mono-laurate, a tetra-glycerol = JIRAU rate, Tetra-glycerol = Monopalmitate, tetra-glycerol = dipalmitate, Tetra-glycerol = Monostearate, tetra-glycerol = distearate, Tetra-glycerol = Monooleate, tetra-glycerol = dioleate, Tetra-glycerol = A MONORINO rate, a tetra-glycerol = JIRINO rate, Tetra-glycerol = Mono-triricinolate, a tetra-glycerol = JIRISHINO rate, Tetra-glycerol = Mono-behenate, tetra-glycerol = dibehenate, PENTA glycerol = Mono-laurate, a PENTA glycerol = JIRAU rate, PENTA glycerol = Monopalmitate, PENTA glycerol = dipalmitate, PENTA glycerol = Monostearate, PENTA glycerol = distearate, PENTA glycerol = Monooleate, PENTA glycerol = dioleate, PENTA glycerol = A MONORINO rate, a PENTA glycerol = JIRINO rate, PENTA glycerol = Mono-triricinolate, a PENTA glycerol = JIRISHINO rate, PENTA glycerol = Mono-behenate, PENTA glycerol = dibehenate, Deca glycerol = Mono-laurate, a deca glycerol = JIRAU rate, Deca glycerol = Trilaurate, deca glycerol = monopalmitate, Deca glycerol = Dipalmitate, deca glycerol = tripalmitate, Deca glycerol = Monostearate, deca glycerol = distearate, Deca glycerol = Tristearate, decaglycerin monooleate, Deca glycerol = Dioleate, deca glycerol = trio REETO, Deca glycerol = A MONORINO rate, a deca glycerol = JIRINO rate, Deca glycerol = A TORIRINO rate, deca glycerol = mono-triricinolate, Deca glycerol = A JIRISHINO rate, a deca glycerol = TORIRISHINO rate, Deca glycerol = Mono-behenate, deca glycerol = dibehenate, Deca glycerol = Tribehenate, deca glycerol = monoisostearate, Deca glycerol = Sesqui-isostearate, deca glycerol = diisostearate, Deca glycerol = TORIISO stearate, deca glycerol = monochrome (12-hydroxy) stearate, Deca glycerol = the fatty acid ester of polyglycerin, such as JI (12-hydroxy) stearate and deca glycerol = Tori (12-hydroxy) stearate, [0013] (iii) Propylene glycol = [Mono-laurate, propylene glycol = monopalmitate,] Propylene glycol = Monostearate, propylene glycol = monooleate, Propylene glycol = A MONORINO rate, propylene glycol = mono-triricinolate, Propylene glycol = The fatty acid ester of propylene glycols, such as monoisostearate and propylene glycol = monochrome (12-hydroxy) stearate, (iv) Sorbitan monolaurate, a sorbitan = JIRAU rate, Sorbitan monopalmitate, sorbitan = dipalmitate, sorbitan = monostearate, Sorbitan = Distearate, sorbitan monooleate, sorbitan = dioleate, Sorbitan = A MONORINO rate, a sorbitan = JIRINO rate, sorbitan = mono-triricinolate, Sorbitan = A JIRISHINO rate, sorbitan = mono-behenate, sorbitan dibehenate, The fatty acid ester of sorbitan, such as sorbitan monoisostearate, sorbitan = diisostearate, sorbitan = monochrome (12-hydroxy) stearate, and sorbitan = JI (12-hydroxy) stearate, [0014] (v) Cane sugar = [Mono-laurate, a cane-sugar = JIRAU rate, cane-sugar = monopalmitate,] Cane sugar = Dipalmitate, cane-sugar = monostearate, cane-sugar = distearate, Cane sugar = Monooleate, cane-sugar = dioleate, a cane-sugar = MONORINO rate, Cane sugar = A JIRINO rate, cane-sugar = mono-triricinolate, a cane-sugar = JIRISHINO rate, Cane sugar = lecithin, such as fatty acid ester of cane sugars, such as monoisostearate, cane-sugar = diisostearate, cane-sugar = monochrome (12-hydroxy) stearate, and cane-sugar = JI (12-hydroxy) steer **-TO, and (vi) soybean-oil lecithin, etc. is mentioned. These may use one sort independently and may use it combining two or more sorts.
[0015] In this invention, when using the mixture of the water-soluble compound which can carry out hydrogen bond to water and water in multiplex as said drainage system medium, as for the content of the water in this mixture, it is desirable that it is 10 % of

the weight or more. It is difficult for the content of water to make polyol and a fatty-acid-ester system emulsifier a micell dissolution condition at less than 10 % of the weight, a desired micellar solution is hard to be obtained, and there is a possibility that quick and sufficient adsorption on a propolis original lump front face may not be performed. Consequently, in the extract processing by the ethyl alcohol in degree process, since amelioration of the affinity of the improvement in extraction efficiency and water which a micell catalyst device is not fully demonstrated by the REBINDA effectiveness list, but may be satisfied cannot be achieved, it is not desirable.

[0016] Moreover, in preparation of a micellar solution, it is good 0.1 - 20 weight section and to dissolve preferably per said drainage system medium 100 weight section, and polyol and a fatty-acid-ester system emulsifier at a rate of 1 - 5 weight section. Since there will be a possibility that a micell gestalt may change and it will become inadequate [all] sticking to a propolis original lump front face if it is hard to carry out the micell dissolution of this emulsifier under in the 0.1 weight section and the amount of dissolutions of polyol and a fatty-acid-ester system emulsifier exceeds 20 weight sections, it is not desirable.

[0017] As for this micellar solution, in this invention, it is advantageous to make it contact at a rate of 15 weight sections at least to the propolis original lump 100 weight section. Infiltration on a propolis original lump front face and adsorption are fully hard to perform the amount of a micellar solution under in 15 weight sections. This infiltration and adsorption actuation are usually performed using Brabender, a kneader, etc., and 30-100 degrees C is usually preferably selected from the point of workability in 50-70 degrees C as temperature in that case.

[0018] Next, after doing in this way and contacting said micellar solution to a propolis original lump, extract processing by ethyl alcohol is performed. per propolis original lump 100 weight section from the point which extracts smoothly and raises a quantitative formula in this extract processing, and ethyl alcohol — usually — the 100 - 1000 weight section — the 200 - 500 weight section comes out comparatively preferably, and it uses.

[0019] In this invention, since a micell catalyst device works automatically also in the state of standing, an extract is also made quickly, but if the whole system is heated or it shakes or mixes [stirring], the REBINDA effectiveness will be demonstrated better, consequently extract processing will be accelerated further.

[0020] In this invention, after doing in this way and performing extract processing, solid liquid separation is usually performed to the bottom of ordinary temperature, and it separates into an alcoholic solution and insoluble propolis residue (extract residue). There is especially no limit as the approach of solid liquid separation, and filtration, a centrifuge method, etc. using a well-known approach, for example, the decantation method, a filter paper, a filter cloth, a wire gauze, etc. can be used conventionally. It is offered as solid-like propolis original object workpiece with which the alcoholic above-mentioned solution adsorbs as a water compatibility alcoholic solution mold propolis food constituent, and extract residue comes to adsorb this constituent.

[0021] This water compatibility alcoholic solution mold propolis food constituent makes the amount of the ethyl alcohol in it fluctuate on the occasion of commercial production, and can be adjusted to suitable concentration and viscosity. Thus, water compatibility is good and the alcoholic solution mold propolis food constituent which is easy to eat is obtained with sufficient extraction efficiency.

[0022] Moreover, solid-like propolis original object workpiece is variously useful as a functional product as an ingredient, a plant growth regulator, etc. of the application of makeup, for example, the charge for the skins. In addition, generally in the manufacture approach of the propolis food constituent by the conventional simple ethyl alcohol extract, extract residue is disposed of as industrial waste.

[Translation done.]

EXAMPLE

[Example] Next, although an example explains this invention to a detail further, this invention is not limited at all by these examples.
[0024] After teaching the propolis original lump 100 weight section from Brazil to example 1 Brabender, the micellar solution which consists of the water 100 weight section, the glycerol 10 weight section, and the tetra-glycerol = mono-laurate 0.4 weight section is added, under ordinary temperature, by 1000rpm, stirring was performed for 15 minutes and mutual contact was carried out. Subsequently, this contact processing object whole quantity was moved to the Erlenmeyer flask with ground-in stopper, the ethyl alcohol 300 weight section adjusted to 20 degrees C at this was added, and standing was carried out for two weeks in 20-degree-C thermostatic chamber. Then, while processing for 30 minutes by 1000rpm under ordinary temperature, extracting the upper supernatant and obtaining the water compatibility alcoholic solution mold propolis food constituent of this invention with a centrifugal separator, the solid-like residue in the condition of having adsorbed it was extracted from the lower layer.
[0025] The propolis original lump 100 weight section from Brazil was put into the example of comparison 1 Erlenmeyer flask with ground-in stopper, the ethyl alcohol 300 weight section subsequently to 20 degrees C adjusted was added, and standing was carried out for two weeks in 20-degree-C thermostatic chamber. Then, while processing for 30 minutes by 1000rpm, extracting the supernatant and obtaining an alcoholic solution mold propolis food constituent under ordinary temperature with a centrifugal separator, lower layer solid-like residue was extracted.
[0026] About the propolis food constituent obtained in the example 1 and the example 1 of a comparison, the total amount of flavonoid was measured and the propolis quantitative formula was computed. Moreover, it put and water compatibility was checked, after adding to underwater [which was adjusted to 20 degrees C / 100g] and mixing 5g of each propolis food constituent for 1 minute. A result is shown in Table 1.
[0027]
[Table 1]

表1

試　料	プロポリス成分含有量（重量%）	水 親 和 性		
		混合直後	1日静置後	5日静置後
実施例1のﾌﾟﾛﾎﾟﾘｽ食品組成物	２４．５	均一乳化状態	均一乳化状態	均一乳化状態
比較例1のﾌﾟﾛﾎﾟﾘｽ食品組成物	１９．７	凝集分離（液白濁）	凝集分離（液半透明）	凝集分離（液透明）

[0028] As for the propolis food constituent of an example 1, it turns out that a sampling volume also has water compatibility a good top, and tends to eat it so that clearly from Table 1.
[0029] The micellar solution which consists of the sorbitan monolaurate 0.3 weight section, the cane-sugar = monooleate 0.2 weight section, and the water 50 weight section is added, after teaching the propolis original lump 100 weight section from Australia to the 4 opening flask equipped with measuring tubing linked to example 2 agitator, a thermometer, a gas inhalant canal, and a capacitor, at 50-60 degrees C, stirring is performed for 2 hours, it contacted and it was mixed. Subsequently, the ethyl alcohol 800 weight section is added to this, and stirring is performed at 50-60 degrees C for 1 hour, and a shaking and after making it mix, passed nitrogen gas, the ethyl alcohol of the 500 weight sections was made to distill off, stirring was stopped, and it was made to cool to ordinary temperature. Then, while filtering under ordinary temperature using the No. 2 filter paper, extracting the filtrate and obtaining the water compatibility alcoholic solution mold propolis food constituent of this invention, the solid-like residue in the condition of having adsorbed it was extracted from the filter paper.
[0030] The propolis original lump 100 weight section from Australia is taught, the ethyl alcohol 800 weight section is added further, stirring is performed at 50-60 degrees C for 1 hour, a shaking and after making it mix, nitrogen gas was passed in the 4 opening flask equipped with measuring tubing linked to example of comparison 2 agitator, a thermometer, a gas inhalant canal, and a capacitor, and it was made to distill off the ethyl alcohol of the 500 weight sections. Subsequently, add the sorbitan monolaurate 0.3 weight section and the cane-sugar = monooleate 0.2 weight section to this, it was made to dissolve in it in 50-60 degrees C, stirring was stopped, and it was made to cool to ordinary temperature. Then, while filtering under ordinary temperature using the No. 2 filter paper, extracting the filtrate and obtaining an alcoholic solution mold propolis food constituent, solid-like residue was extracted from the filter paper.
[0031] While measuring the propolis quantitative formula like the example 1, respectively about the propolis food constituent obtained in the example 2 and the example 2 of a comparison, the water compatibility trial was performed. A result is shown in Table 2.
[0032]
[Table 2]

表2

試　料	プロポリス成分含有量（重量%）	水親和性		
		混合直後	1日静置後	5日静置後
実施例2のﾌﾟﾛﾎﾟﾘｽ食品組成物	36．5	均一乳化状態	均一乳化状態	均一乳化状態
比較例2のﾌﾟﾛﾎﾟﾘｽ食品組成物	28．9	均一分散状態	上層に分離液が形成	下層に沈殿が形成

[0033] In the example 2 which is carrying out extract processing of the solution which comes to carry out the micell dissolution of the emulsifier component into a drainage system medium with ethyl alcohol since a propolis original lump is contacted beforehand , an emulsifier component is understood that the top where the extraction efficiency of a propolis component is high and water compatibility are good compared with the example 2 of a comparison which is carrying out extract processing by the system which carries out the simple dissolution in ethyl alcohol so that clearly from Table 2 .

[0034] The propolis original lump of a class and an amount which shows in Table 3 and 4 according to the approach of three to example 10 example 1, or an example 2, By performing extract processing by ethyl alcohol after contact processing on the conditions shown in Table 3 and 4 using a drainage system medium, and polyol and a fatty-acid-ester system emulsifier, and subsequently performing after treatment While obtaining the water compatibility alcoholic solution mold propolis food constituent, the solid-like residue in the condition of having adsorbed it was extracted.

[0035]

[Table 3]

表3

実施例	プロポリス原塊	媒　体	ポリオール・脂肪酸エステル系乳化剤	エチルアルコール	製造方法
3	ブラジル産プロポリス原塊 100重量部	水　15重量部	ｸﾞﾘｾﾘﾝ=ﾓﾉﾗｳﾚｰﾄ 0.05重量部 ﾍﾟﾝﾀｸﾞﾘｾﾘﾝ=ﾓﾉｽﾃｱﾚｰﾄ 0.1重量部	500重量部投入し、常温で48時間静置、200重量部を系外に除去した。	実施例1の製造方法に準ずる
4	米国産プロポリス原塊 100重量部	水　15.5重量部 D-ｿﾙﾋﾞｯﾄ 3重量部 ｸﾞﾙｺﾉﾃﾞﾙﾗｸﾄﾝ 1重量部 ﾌﾟﾛﾋﾟﾚﾝｸﾞﾘｺｰﾙ 0.5重量部	ﾌﾟﾛﾋﾟﾚﾝｸﾞﾘｺｰﾙ-ﾓﾉﾗｳﾚｰﾄ 0.01重量部 ｼﾞｸﾞﾘｾﾘﾝ=ﾓﾉﾘｼﾉﾚｰﾄ 0.01重量部	300重量部投入し、常温で72時間静置した。	同　上
5	ドイツ産プロポリス原塊 100重量部	水　3重量部 ｸｴﾝ酸　1重量部 ｸﾞﾘｾﾘﾝ 26重量部	ｼｮ糖=ﾓﾉﾊﾟﾙﾐﾃｰﾄ 2重量部 ﾃﾞｶｸﾞﾘｾﾘﾝ=ﾓﾉﾘﾉﾚｰﾄ 4重量部	300重量部投入し、40℃で15時間静置した。	同　上
6	中国産プロポリス原塊 100重量部	水　15重量部 D-ｷｼﾛｰｽ 4重量部 ｸﾞﾘｼﾝ 1重量部	ｿﾙﾋﾞﾀﾝ=ﾓﾉｵﾚｴｰﾄ 0.04重量部 ﾃﾄﾗｸﾞﾘｾﾘﾝ=ﾓﾉ12-ﾋﾄﾞﾛｷｼｽﾃｱﾚｰﾄ 0.04重量部	200重量部投入し、50℃で24時間静置した後、さらに、常温で100重量部投入した。	同　上

[0036]

[Table 4]

表4

実施例	プロポリス原塊	媒体	ポリオール・脂肪酸エステル系乳化剤	エチルアルコール	製造方法
7	米国産プロポリス原塊 100重量部	水 23重量部 リンゴ酸 2重量部	ペンタグリセリン・ジラウレート 0.4重量部 ショ糖・モノステアレート 0.4重量部	300重量部投入し、60℃で2時間、攪拌、混合させた。	実施例2の製造方法に準ずる
8	ドイツ産プロポリス原塊 100重量部	水 30重量部 クエン酸 2重量部 ゲルコン酸 1重量部 グリセリン 7重量部	ショ糖=モノオレエート 3重量部 ペンタグリセリン・モノイソステアレート 4重量部 大豆油レシチン 1重量部	800重量部投入し、常温で24時間攪拌・混合させた後、500重量部を系外に除去した。	同上
9	中国産プロポリス原塊 100重量部	水 5重量部 D-キシロース 5重量部 グリセリン 40重量部	グリセリン=モノリシノレート 0.2重量部 デカグリセリン=ジラウレート 0.2重量部	300重量部投入し、常温で24時間、攪拌・混合させた。	同上
10	オーストラリア産プロポリス原塊 100重量部	水 35重量部 D-ソルビット 12重量部 酒石酸 3重量部	ソルビタン=モノラウレート 0.03重量部 ショ糖=ジラウレート 0.02重量部	300重量部投入し、50℃で3時間、攪拌・混合させた。	同上

[0037] While measuring the propolis quantitative formula like the example 1 about the propolis food constituent obtained in the examples 3-10, respectively, the water compatibility trial was performed. A result is shown in Table 5.
[0038]
[Table 5]

表5

試料	プロポリス成分含有量（重量%）	水親和性		
		混合直後	1日静置後	5日静置後
実施例3のプロポリス食品組成物	17.1	均一乳化状態	均一乳化状態	均一乳化状態
実施例4のプロポリス食品組成物	16.3	同上	同上	同上
実施例5のプロポリス食品組成物	19.2	同上	同上	同上
実施例6のプロポリス食品組成物	16.1	同上	同上	同上
実施例7のプロポリス食品組成物	18.8	同上	同上	同上
実施例8のプロポリス食品組成物	18.9	同上	同上	同上
実施例9のプロポリス食品組成物	17.5	同上	同上	同上
実施例10のプロポリス食品組成物	16.2	同上	同上	同上

[0039] Each propolis food constituent of examples 3-10 has the good extraction efficiency of a propolis component, and the water compatibility made into the purpose also comes out enough, and understands a certain thing for it so that clearly from

Table 5.
[0040] The water 12 weight section, the glycerol 64.7 weight section, the stearin acid 8 weight section, and the sodium-hydroxide 0.3 weight section were taught to the beaker which attached application 1 agitator and the thermometer, and it mixed, heating gradually, and the milky lotion was prepared at 80 degrees C. Subsequently, after lowering an internal temperature to 50 degrees C, the dirt dropping cream of a dispersion mold was produced by supplying the solid-like residue 15 weight section obtained in the example 1, and mixing homogeneity for 30 minutes by 1000rpm.
[0041] In comparison application 1 application 1, the dirt dropping cream of a dispersion mold was produced like the application 1 except having used the solid-like residue obtained in the example 1 of a comparison instead of the solid-like residue obtained in the example 1.
[0042] Standing was carried out to the thermostat which moved 10g of dirt dropping creams of an application 1 and the comparison application 1 at a time to the glass cylinder, respectively, and adjusted them to 25 degrees C for one month, and the stability of dispersion was observed.
[0043] I had the feeling after the dirt dropping effectiveness after having divided into ten persons at a time 20 persons who chose at random about each dirt dropping cream, having the arm and the hand use them after activity termination and having them subsequently wiped off with a cloth towel, a touch rate, and use told on the other hand out of the 18-60-year-old man who is doing the field work about the thing immediately after manufacture in the car garage for working 8 hours for one day. These results are shown in Table 6.
[0044]
[Table 6]

表6

試　料	ディスパージョン安定性（25℃、1ヶ月後）	汚れ落とし効果　1)	肌触わり　2)	使用後の感覚　3)
応用例1の汚れ落としクリーム	均一状態	○	○	○
比較応用例1の汚れ落としクリーム	底部に沈殿物形成	△	×	△

[0045] (Note)
1) The inside of the thing **:10 person examiner who eight or more persons' person accepted was good among the valuation-basis O:10 person [of the dirt dropping effectiveness] examiner, Those who admited being good among the thing x:10 person examiner who 5-7 persons accepted was good The inside of a valuation-basis O:10 person [of four or less persons' thing 2 touch rate] examiner, The inside of the thing x:10 person examiner who 5-7 persons' person accepted was good among the thing **:10 person examiner who eight or more persons' person accepted was good, The inside of an evaluation O:10 person [of the feeling after the thing 3 use which six or more persons' person accepted to be a poor touch rate] examiner, Those who conceded carried out gently among the thing **:10 person examiner who eight or more persons' person accepted carried out gently, and the thing x:10 person examiner who 5-7 persons accepted carried out gently are four or less persons' things [0046]. After sprinkling 2kg of solid-like residue obtained in the example 2 on the average to the place to 20cm of surfaces with application 2 above sea level of 300m of Kanto loam layer Hataji 12m2, 50 seedlings of lavender were planted and it was made to grow them for summer three months.
[0047] After sprinkling 2kg of solid-like residue obtained in the example 2 of a comparison on the average to the place to 20cm of surfaces of Kanto loam layer Hataji 12m2 which adjoin the location in comparison application 2 application 2, 50 seedlings of lavender were planted and it was grown for the same stage three months with the application 2. Moreover, 50 seedlings of lavender were planted in Kanto loam layer Hataji 12m2 which adjoin an application 2 and the comparison application 2, and it was made to grow for three months as it is at a coincidence term. Each growth situation is summarized and it is shown in Table 7.
[0048]
[Table 7]

表7

試　料	頭頂の花部までの長さ			茎部の状態　1)
	45cmより高いもの	35～45cmのもの	35cm未満のもの	
無処理地区のラベンダー	0本／50本	12本／50本	38本／50本	C
応用例2のラベンダー	23本／50本	26本／50本	1本／50本	A
比較応用例2のラベンダー	3本／50本	28本／50本	19本／50本	B

[0049] (Note)
1) The thing C to which 30-39 are strongly extended towards the upper part among A:50 valuation bases of the condition of a scapus among B:50 things to which 40 or more are strongly extended towards the upper part : it turns out that the growth situation of the lavender in an application 2 is good so that clearly [what is strongly extended towards the upper part] from 29 or less thing tables 7.

[Translation done.]

(A)-(4)JP 2002-84992.A

(19)日本国特許庁（ＪＰ）　(12)公開特許公報（A）　(11)特許出願公開番号
特開2002－84992
（Ｐ２００２－８４９９２Ａ）
(43)公開日　平成14年３月26日(2002.3.26)

(51)Int.Cl.[7]	識別記号	Ｆ Ｉ		テーマコード（参考）
A23L 1/076		A23L 1/076		4B018
1/30		1/30	A	4B041
A61K 7/00		A61K 7/00	K	4C083

審査請求　未請求　請求項の数４　ＯＬ　（全８頁）

(21)出願番号　特願2000－282859(Ｐ2000－282859)

(22)出願日　平成12年９月19日(2000.9.19)

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最終頁に続く

(54)【発明の名称】溶液型プロポリス食品組成物および固形状プロポリス原体加工物の製造方法

(57)【要約】
【課題】　食しやすい水親和性を有するアルコール溶液型のプロポリス食品組成物を抽出効率よく製造すると同時に、各種用途に有用な固形状プロポリス原体加工物の製造方法を提供する。
【解決手段】　プロポリス原塊に対し、水系媒体とポリオール・脂肪酸エステル系乳化剤を含むミセル溶液を接触させたのち、エチルアルコールによる抽出処理を施し、次いで固液分離することにより、水親和性アルコール溶液型プロポリス食品組成物およびそれを吸着してなる固形状プロポリス原体加工物を製造する。

1

【特許請求の範囲】

【請求項１】　プロポリス原塊に対し、水系媒体とポリオール・脂肪酸エステル系乳化剤を含むミセル溶液を接触させたのち、エチルアルコールによる抽出処理を施し、次いで固液分離することを特徴とする、水親和性アルコール溶液型プロポリス食品組成物およびそれを吸着してなる固形状プロポリス原体加工物の製造方法。

【請求項２】　水系媒体が、水単独または水および水と多重的に水素結合し得る水溶性化合物の混合物である請求項１に記載の方法。

【請求項３】　固形状プロポリス原体加工物が、皮膚用化粧料の材料として用いられる請求項１または２に記載の方法。

【請求項４】　固形状プロポリス原体加工物が、植物生長調節剤として用いられる請求項１または２に記載の方法。

【発明の詳細な説明】

【０００１】

【発明の属する技術分野】本発明は、溶液型プロポリス食品組成物および固形状プロポリス原体加工物の製造方法に関する。さらに詳しくは、本発明は、食しやすい水親和性を有するアルコール溶液型のプロポリス食品組成物を、抽出効率よく製造すると同時に、上記プロポリス食品組成物を吸着し、各種用途に有用な固形状プロポリス原体加工物をも製造する工業的に有利な方法に関するものである。

【０００２】

【従来の技術】古くから天然の抗菌剤として知られるプロポリスは、ミツバチが樹木の特定部位、主として新芽や蕾および樹皮から採取したガム質、樹液、植物色素系の物質並びに香油などの集合体に、ミツバチ自身の分泌物、蜂ろうなどを混合して作られた樹脂状の固形物質である。これを食する場合には、そのままでは硬くて不適であるために、通常エチルアルコールや液化二酸化炭素や水、液状多糖類などを用いる溶液抽出によって、溶液状食品とし、さらに媒体で希釈できるような形態にして、供されてきた。

【０００３】これらの抽出方法の中で、水抽出法や液状多糖類抽出法は、成分抽出量が低いという問題があるし、液化二酸化炭素抽出法では、抽出後に固化した成分を他の媒体中に再溶解させて、食しやすい形態へ変換する工程が必要となり、操作が煩雑であるという問題があった。このような事情から、成分抽出量が多く、しかも操作が簡単なエチルアルコールによる抽出法が、従来最も多く行われてきた。

【０００４】ところで、プロポリス食品は、それを食する際には、水で希釈して口中に入れるので、水に溶解若しくは分散する性質を有することが好ましく、その意味では、エチルアルコール抽出法によって得られたプロポリス食品は、非水溶性であるために、水で希釈すると成分が析出して、容器に付着したままになったり、スムーズに口、食道から胃や腸へ移行されないなど、好ましくない事態を招来し、その改良が望まれていた。

2

【０００５】そこで、本発明者らは、エチルアルコール抽出プロポリス食品の水との親和性の向上を図るために、先に、ポリオール・脂肪酸エステル系乳化剤と水系媒体を含むミセル溶液に、エチルアルコール抽出プロポリス食品を接触、混合させることにより、抽出成分の表面を改質し、水分散性を呈するプロポリス食品組成物を製造する方法（特公平４－６６５４４号公報）、エチルアルコール抽出プロポリス食品の中にサポニンを添加する方法（特開平６－１９７７３４号公報）、エチルアルコール抽出プロポリス食品を穀物タンパク質部分分解物溶液と接触させて、水分散性を付与する方法（特開平９－７５０１８号公報）などを見出した。

【０００６】一方、本発明者らは、始めから水との親和性の良好なプロポリス食品組成物を製造するために、前述の特公平４－６６５４４号公報におけるもう１つの発明において、プロポリス原塊そのものをポリオール・脂肪酸エステル系乳化剤の水系媒体ミセルと接触させる製造方法を示し、水可溶化型のプロポリス食品組成物を一工程で作製することに成功した。しかしながら、この方法は、水との親和性は極めて満足し得るものの、成分抽出量の点で、エチルアルコール抽出プロポリス食品に比べて劣るという問題があった。また、従来の単純エチルアルコール抽出によるプロポリス食品組成物の製造方法においては、不溶のプロポリス残渣（抽出残渣）は、一般に産業廃棄物として処分されており、その有効利用の開発が望まれていた。

【０００７】

【発明が解決しようとする課題】本発明は、このような事情のもとで、プロポリス原塊からの一貫工程によって、抽出効率の向上と水との親和力の改良の両方を同時に図ることのできるアルコール溶液型のプロポリス食品組成物と、機能性製品として種々の用途に有用な不溶のプロポリス残渣（抽出残渣）の製造方法を提供することを目的とするものである。

【０００８】

【課題を解決するための手段】本発明者らは、前記目的を達成するために鋭意研究を重ねた結果、まず、プロポリス原塊を、水系媒体とポリオール・脂肪酸エステル系乳化剤を含むミセル溶液と接触させて十分に浸潤、吸着させたのち、エチルアルコールにより抽出処理を行うことにより、エチルアルコールのプロポリス原塊内への浸透作用および成分の抽出、溶解作用が向上し、しかも固液分離により抽出残渣を除去したアルコール溶液は、従来のエチルアルコール抽出プロポリス食品組成物に対して、後添加で乳化剤成分を含有させたものよりも安定な水親和性を有するアルコール溶液型プロポリス食品組成物となること、そして上記抽出残渣は該組成物を吸着

し、機能性製品として種々の用途に有用な固形状プロポリス原体加工物となることを見出した。本発明は、これらの知見に基づいて完成したものである。

【０００９】すなわち、本発明は、プロポリス原塊に対し、水系媒体とポリオール・脂肪酸エステル系乳化剤を含むミセル溶液を接触させたのち、エチルアルコールによる抽出処理を施し、次いで固液分離することを特徴とする、水親和性アルコール溶液型プロポリス食品組成物およびそれを吸着してなる固形状プロポリス原体加工物の製造方法を提供するものである。

【００１０】

【発明の実施の形態】本発明の方法における原料のプロポリス原塊としては特に制限されず、いかなる由来のものであってもよく、例えば、ブラジル産、米国産、ドイツ産、中国産、オーストラリア産など、いずれも用いることができる。

【００１１】本発明の方法においては、まず、上記プロポリス原塊に対し、水系媒体とポリオール・脂肪酸エステル系乳化剤を含むミセル溶液を接触させる。上記ミセル溶液における水系媒体としては、水単独であってもよいし、水および水と多重的に水素結合し得る水溶性化合物の混合物であってもよい。ここで、水と多重的に水素結合し得る水溶性化合物の例としては、プロピレングリコール、グリセリン、Ｄ－キシロース、Ｄ－ソルビット、クエン酸、リンゴ酸、コハク酸、フマル酸、酒石酸、グルコン酸、グルコノデルタラクトン、グリシンなどを挙げることができる。これらは１種を単独で用いてもよいし、２種以上を組み合わせて用いてもよい。

【００１２】一方、ポリオール・脂肪酸エステル系乳化剤としては、例えば(i) グリセリン＝モノラウレート、グリセリン＝モノパルミテート、グリセリン＝モノステアレート、グリセリン＝モノオレエート、グリセリン＝モノリノレート、グリセリン＝モノリシノレートなどのグリセリンの脂肪酸エステル、(ii)ジグリセリン＝モノラウレート、ジグリセリン＝モノパルミテート、ジグリセリン＝モノステアレート、ジグリセリン＝モノオレエート、ジグリセリン＝モノリノレート、ジグリセリン＝モノリシノレート、テトラグリセリン＝モノラウレート、テトラグリセリン＝ジラウレート、テトラグリセリン＝モノパルミテート、テトラグリセリン＝ジパルミテート、テトラグリセリン＝モノステアレート、テトラグリセリン＝ジステアレート、テトラグリセリン＝モノオレエート、テトラグリセリン＝ジオレエート、テトラグリセリン＝モノリノレート、テトラグリセリン＝ジリノレート、テトラグリセリン＝モノリシノレート、テトラグリセリン＝ジリシノレート、テトラグリセリン＝モノベヘネート、テトラグリセリン＝ジベヘネート、ペンタグリセリン＝モノラウレート、ペンタグリセリン＝ジラウレート、ペンタグリセリン＝モノパルミテート、ペンタグリセリン＝ジパルミテート、ペンタグリセリン＝モノステアレート、ペンタグリセリン＝ジステアレート、ペンタグリセリン＝モノオレエート、ペンタグリセリン＝ジオレエート、ペンタグリセリン＝モノリノレート、ペンタグリセリン＝ジリノレート、ペンタグリセリン＝モノリシノレート、ペンタグリセリン＝ジリシノレート、ペンタグリセリン＝モノベヘネート、ペンタグリセリン＝ジベヘネート、デカグリセリン＝モノラウレート、デカグリセリン＝ジラウレート、デカグリセリン＝トリラウレート、デカグリセリン＝モノパルミテート、デカグリセリン＝ジパルミテート、デカグリセリン＝トリパルミテート、デカグリセリン＝モノステアレート、デカグリセリン＝ジステアレート、デカグリセリン＝トリステアレート、デカグリセリン＝モノオレエート、デカグリセリン＝ジオレエート、デカグリセリン＝トリオレエート、デカグリセリン＝モノリノレート、デカグリセリン＝ジリノレート、デカグリセリン＝トリリノレート、デカグリセリン＝モノリシノレート、デカグリセリン＝ジリシノレート、デカグリセリン＝トリリシノレート、デカグリセリン＝モノベヘネート、デカグリセリン＝ジベヘネート、デカグリセリン＝トリベヘネート、デカグリセリン＝モノイソステアレート、デカグリセリン＝セスキイソステアレート、デカグリセリン＝ジイソステアレート、デカグリセリン＝トリイソステアレート、デカグリセリン＝モノ（１２－ヒドロキシ）ステアレート、デカグリセリン＝ジ（１２－ヒドロキシ）ステアレート、デカグリセリン＝トリ（１２－ヒドロキシ）ステアレートなどのポリグリセリンの脂肪酸エステル、

【００１３】(iii) プロピレングリコール＝モノラウレート、プロピレングリコール＝モノパルミテート、プロピレングリコール＝モノステアレート、プロピレングリコール＝モノオレエート、プロピレングリコール＝モノリノレート、プロピレングリコール＝モノリシノレート、プロピレングリコール＝モノイソステアレート、プロピレングリコール＝モノ（１２－ヒドロキシ）ステアレートなどのプロピレングリコールの脂肪酸エステル、(iv)ソルビタン＝モノラウレート、ソルビタン＝ジラウレート、ソルビタン＝モノパルミテート、ソルビタン＝ジパルミテート、ソルビタン＝モノステアレート、ソルビタン＝ジステアレート、ソルビタン＝モノオレエート、ソルビタン＝ジオレエート、ソルビタン＝モノリノレート、ソルビタン＝ジリノレート、ソルビタン＝モノリシノレート、ソルビタン＝ジリシノレート、ソルビタン＝モノベヘネート、ソルビタン＝ジベヘネート、ソルビタン＝モノイソステアレート、ソルビタン＝ジイソステアレート、ソルビタン＝モノ（１２－ヒドロキシ）ステアレート、ソルビタン＝ジ（１２－ヒドロキシ）ステアレートなどのソルビタンの脂肪酸エステル、

【００１４】(v) ショ糖＝モノラウレート、ショ糖＝ジラウレート、ショ糖＝モノパルミテート、ショ糖＝ジパルミテート、ショ糖＝モノステアレート、ショ糖＝ジス

テアレート、ショ糖＝モノオレエート、ショ糖＝ジオレエート、ショ糖＝モノリノレート、ショ糖＝ジリノレート、ショ糖＝モノリシノレート、ショ糖＝ジリシノレート、ショ糖＝モノイソステアレート、ショ糖＝ジイソステアレート、ショ糖＝モノ（１２－ヒドロキシ）ステアレート、ショ糖＝ジ（１２－ヒドロキシ）ステアれートなどのショ糖の脂肪酸エステル、(vi)大豆油レシチンなどのレシチン、などが挙げられる。これらは１種を単独で用いてもよいし、２種以上を組み合わせて用いてもよい。

【００１５】本発明においては、前記水系媒体として水および水と多重的に水素結合し得る水溶性化合物の混合物を用いる場合には、この混合物における水の含有量は、１０重量％以上であることが望ましい。水の含有量が１０重量％未満ではポリオール・脂肪酸エステル系乳化剤をミセル溶解状態とすることが困難であって、所望のミセル溶液が得られにくく、プロポリス原塊表面への迅速かつ十分な吸着が行われないおそれがある。その結果、次工程におけるエチルアルコールによる抽出処理において、レビンダー効果並びにミセル触媒機構が十分に発揮されず、満足し得る抽出効率の向上と水との親和力の改良を果すことができないので、好ましくない。

【００１６】また、ミセル溶液の調製においては、前記水系媒体１００重量部当たり、ポリオール・脂肪酸エステル系乳化剤を、０．１～２０重量部、好ましくは１～５重量部の割合で溶解させるのがよい。ポリオール・脂肪酸エステル系乳化剤の溶解量が０．１重量部未満では該乳化剤がミセル溶解しにくいし、２０重量部を超えるとミセル形態が変化するおそれがあり、いずれもプロポリス原塊表面への吸着が不十分となるので、好ましくない。

【００１７】本発明においては、このミセル溶液は、プロポリス原塊１００重量部に対し、少なくとも１５重量部の割合で接触させるのが有利である。ミセル溶液の量が１５重量部未満ではプロポリス原塊表面への浸潤、吸着が十分に行われにくい。この浸潤、吸着操作は、通常ブラベンダーやニーダなどを使用して行われ、また、その際の温度としては、作業性の点から、通常３０～１００℃、好ましくは５０～７０℃の範囲で選定される。

【００１８】次に、このようにして、プロポリス原塊に対し、前記ミセル溶液を接触させたのち、エチルアルコールによる抽出処理を施す。この抽出処理においては、抽出を円滑に行い、かつ成分含有量を高める点から、プロポリス原塊１００重量部当たり、エチルアルコールを、通常１００～１０００重量部、好ましくは２００～５００重量部の割合で用いる。

【００１９】本発明においては、静置状態でも自然にミセル触媒機構が働くので、抽出も迅速になされるが、系全体を加熱したり振とうまたは撹拌混合させると、レビンダー効果がより良く発揮され、その結果、抽出処理が一層加速される。

【００２０】本発明においては、このようにして抽出処理を施したのち、通常常温下において固液分離を行い、アルコール溶液と不溶のプロポリス残渣（抽出残渣）とに分離する。固液分離の方法としては特に制限はなく、従来公知の方法、例えばデカンテーション法、ろ紙、ろ布、金網などを用いるろ過法、遠心分離法などを用いることができる。上記アルコール溶液は、水親和性アルコール溶液型プロポリス食品組成物として、また抽出残渣は、該組成物を吸着してなる固形状プロポリス原体加工物として供せられる。

【００２１】該水親和性アルコール溶液型プロポリス食品組成物は、製品化に際してはその中のエチルアルコールの量を増減させて、適当な濃度および粘度に調整することができる。このようにして、水親和性が良好で、食しやすいアルコール溶液型プロポリス食品組成物が、抽出効率よく得られる。

【００２２】また、固形状プロポリス原体加工物は、機能性製品として種々その用途、例えば皮膚用化粧料の材料や植物生長調節剤などとして有用である。なお、従来の単純エチルアルコール抽出によるプロポリス食品組成物の製造方法においては、抽出残渣は、一般に産業廃棄物として処分されていたものである。

【００２３】

【実施例】次に、本発明を実施例により、さらに詳細に説明するが、本発明は、これらの例によってなんら限定されるものではない。

【００２４】実施例１

ブラベンダーに、ブラジル産プロポリス原塊１００重量部を仕込んだのち、水１００重量部、グリセリン１０重量部およびテトラグリセリン＝モノラウレート０．４重量部からなるミセル溶液を加え、常温下、１０００ｒｐｍで１５分間攪拌を行い、相互接触させた。次いで、この接触処理物全量を共栓付き三角フラスコに移し取り、これに２０℃に調整したエチルアルコール３００重量部を加え、２０℃恒温室にて２週間静置させた。その後、遠心分離機により、常温下に１０００ｒｐｍで３０分間処理し、上層の上澄み液を採取して、本発明の水親和性アルコール溶液型プロポリス食品組成物を得ると共に、それを吸着した状態にある固形状残渣を下層より採取した。

【００２５】比較例１

共栓付き三角フラスコにブラジル産プロポリス原塊１００重量部を入れ、次いで２０℃に調整したエチルアルコール３００重量部を加え、２０℃恒温室にて２週間静置させた。その後、遠心分離機により、常温下に１０００ｒｐｍで３０分間処理して上澄み液を採取し、アルコール溶液型プロポリス食品組成物を得ると共に、下層の固形状残渣を採取した。

【００２６】実施例１および比較例１で得られたプロポ

リス食品組成物について、総フラボノイド量を測定し、プロポリス成分含有量を算出した。また、それぞれのプロポリス食品組成物５ｇを２０℃に調整した１００ｇの水中に添加して１分間混合させたのち、静置し、水親和性の確認を行った。結果を表１に示す。

【００２７】

【表１】

表１

試　料	プロポリス成分含有量（重量%）	水親和性		
		混合直後	１日静置後	５日静置後
実施例１のﾌﾟﾛﾎﾟﾘｽ食品組成物	２４．５	均一乳化状態	均一乳化状態	均一乳化状態
比較例１のﾌﾟﾛﾎﾟﾘｽ食品組成物	１９．７	凝集分離（液白濁）	凝集分離（液半透明）	凝集分離（液透明）

【００２８】表１から明らかなように、実施例１のプロポリス食品組成物は、抽出量も良好である上、水親和性を有し、食しやすいことが分かる。

【００２９】実施例２

撹拌機、温度計、気体流入管およびコンデンサーに接続する検量管を備えた四つ口フラスコに、オーストラリア産プロポリス原塊１００重量部を仕込んだのち、ソルビタン＝モノラウレート０．３重量部、ショ糖＝モノオレエート０．２重量部および水５０重量部からなるミセル溶液を加え、５０～６０℃で２時間撹拌を行い、接触、混合させた。次いで、これにエチルアルコール８００重量部を加え、５０～６０℃で１時間撹拌を行い、振とう、混合させたのち、窒素ガスを流して５００重量部のエチルアルコールを留去させて、撹拌を停止し、常温まで冷却させた。その後、常温下に２号ろ紙を使用してろ過を行い、ろ液を採取して、本発明の水親和性アルコール溶液型プロポリス食品組成物を得ると共に、それを吸着した状態にある固形状残渣をろ紙から採取した。

【００３０】比較例２

撹拌機、温度計、気体流入管およびコンデンサーに接続する検量管を備えた四つ口フラスコに、オーストラリア産プロポリス原塊１００重量部を仕込み、さらにエチルアルコール８００重量部を加え、５０～６０℃で１時間撹拌を行い、振とう、混合させたのち、窒素ガスを流して５００重量部のエチルアルコールを留去させた。次いで、これに５０～６０℃においてソルビタン＝モノラウレート０．３重量部とショ糖＝モノオレエート０．２重量部を加えて溶解させ、撹拌を停止して、常温まで冷却させた。その後、常温下に２号ろ紙を使用してろ過を行い、ろ液を採取して、アルコール溶液型プロポリス食品組成物を得ると共に、ろ紙から固形状残渣を採取した。

【００３１】実施例２および比較例２で得られたプロポリス食品組成物について、それぞれ実施例１と同様にしてプロポリス成分含有量を測定すると共に、水親和性試験を行った。結果を表２に示す。

【００３２】

【表２】

表２

試　料	プロポリス成分含有量（重量%）	水親和性		
		混合直後	１日静置後	５日静置後
実施例２のﾌﾟﾛﾎﾟﾘｽ食品組成物	３６．５	均一乳化状態	均一乳化状態	均一乳化状態
比較例２のﾌﾟﾛﾎﾟﾘｽ食品組成物	２８．９	均一分散状態	上層に分離液が形成	下層に沈殿が形成

【００３３】表２から明らかなように、乳化剤成分を水系媒体中にミセル溶解させてなる溶液を、予めプロポリス原塊に接触させておいてから、エチルアルコールで抽出処理している実施例２では、乳化剤成分をエチルアルコール中に単純溶解させている系で抽出処理している比較例２に比べて、プロポリス成分の抽出効率が高い上、水親和性が良好であることが分かる。

【００３４】実施例３～１０

実施例１または実施例２の方法に準じて、表３および表４に示す種類と量のプロポリス原塊、水系媒体およびポリオール・脂肪酸エステル系乳化剤を用いて接触処理後、表３および表４に示す条件にてエチルアルコールによる抽出処理を行い、次いで後処理を行うことにより、水親和性アルコール溶液型プロポリス食品組成物を得ると共に、それを吸着した状態にある固形状残渣を採取した。

【００３５】

【表３】

表3

実施例	プロポリス原塊	媒　体	ポリオール・脂肪酸エステル系乳化剤	エチルアルコール	製造方法
3	ブラジル産プロポリス原塊 100重量部	水　15重量部	ｸﾞﾘｾﾘﾝ=ﾓﾉﾗｳﾚｰﾄ 0.05重量部 ﾍﾟﾝﾀｸﾞﾘｾﾘﾝ=ﾓﾉｽﾃｱﾚｰﾄ 0.1重量部	500重量部投入し、常温で48時間静置、200重量部を系外に除去した。	実施例１の製造方法に準ずる
4	米国産プロポリス原塊 100重量部	水 15.5重量部 D-ｿﾙﾋﾞｯﾄ 3重量部 ｸﾞﾙｺﾉﾃﾞﾙﾗｸﾄﾝ 1重量部 ﾌﾟﾛﾋﾟﾚﾝｸﾞﾘｺｰﾙ 0.5重量部	ﾌﾟﾛﾋﾟﾚﾝｸﾞﾘｺｰﾙ-ﾓﾉﾗｳﾚｰﾄ 0.01重量部 ｼﾞｸﾞﾘｾﾘﾝ=ﾓﾉﾘｼﾉﾚｰﾄ 0.01重量部	300重量部投入し、常温で72時間静置した。	同　上
5	ドイツ産プロポリス原塊 100重量部	水　3重量部 ｸｴﾝ酸　1重量部 ｸﾞﾘｾﾘﾝ 26重量部	ｼｮ糖=ﾓﾉﾊﾟﾙﾐﾃｰﾄ 2重量部 ﾃﾞｶｸﾞﾘｾﾘﾝ=ﾓﾉﾘﾉﾚｰﾄ 4重量部	300重量部投入し、40℃で15時間静置した。	同　上
6	中国産プロポリス原塊 100重量部	水　15重量部 D-ｷｼﾛｰｽ 4重量部 ｸﾞﾘｼﾝ 1重量部	ｿﾙﾋﾞﾀﾝ=ﾓﾉｵﾚｴｰﾄ 0.04重量部 ﾃﾄﾗｸﾞﾘｾﾘﾝ=ﾓﾉ12-ﾋﾄﾞﾛｷｼｽﾃｱﾚｰﾄ 0.04重量部	200重量部投入し、50℃で24時間静置した後、さらに、常温で100重量部投入した。	同　上

【００３６】　　【表４】

表4

実施例	プロポリス原塊	媒　体	ポリオール・脂肪酸エステル系乳化剤	エチルアルコール	製造方法
7	米国産プロポリス原塊 100重量部	水　23重量部 ﾘﾝｺﾞ酸 2重量部	ﾍﾟﾝﾀｸﾞﾘｾﾘﾝ・ｼﾞﾗｳﾚｰﾄ 0.4重量部 ｼｮ糖・ﾓﾉｽﾃｱﾚｰﾄ 0.4重量部	300重量部投入し、60℃で2時間、撹拌、混合させた。	実施例２の製造方法に準ずる
8	ドイツ産プロポリス原塊 100重量部	水　30重量部 ｸｴﾝ酸　2重量部 ｸﾞﾙｺﾝ酸 1重量部 ｸﾞﾘｾﾘﾝ 7重量部	ｼｮ糖=ﾓﾉｵﾚｴｰﾄ 3重量部 ﾍﾟﾝﾀｸﾞﾘｾﾘﾝ・ﾓﾉｲｿｽﾃｱﾚｰﾄ 4重量部 大豆油ﾚｼﾁﾝ 1重量部	800重量部投入し、常温で24時間撹拌・混合させた後、500重量部を系外に除去した。	同　上
9	中国産プロポリス原塊 100重量部	水　5重量部 D-ｷｼﾛｰｽ 5重量部 ｸﾞﾘｾﾘﾝ 40重量部	ｸﾞﾘｾﾘﾝ=ﾓﾉﾘｼﾉﾚｰﾄ 0.2重量部 ﾃﾞｶｸﾞﾘｾﾘﾝ=ｼﾞﾗｳﾚｰﾄ 0.2重量部	300重量部投入し、常温で24時間、撹拌・混合させた。	同　上
10	ｵｰｽﾄﾗﾘｱ産プロポリス原塊 100重量部	水　35重量部 D-ｿﾙﾋﾞｯﾄ 12重量部 酒石酸 3重量部	ｿﾙﾋﾞﾀﾝ=ﾓﾉﾗｳﾚｰﾄ 0.03重量部 ｼｮ糖=ｼﾞﾗｳﾚｰﾄ 0.02重量部	300重量部投入し、50℃で3時間、撹拌・混合させた。	同　上

【００３７】実施例３～１０で得られたプロポリス食品組成物について、それぞれ実施例１と同様にしてプロポリス成分含有量を測定すると共に、水親和性試験を行った。結果を表５に示す。

【００３８】

【表５】

表5

試　料	プロポリス成分含有量（重量%）	水親和性		
		混合直後	１日静置後	５日静置後
実施例３のﾌﾟﾛﾎﾟﾘｽ食品組成物	１７．１	均一乳化状態	均一乳化状態	均一乳化状態
実施例４のﾌﾟﾛﾎﾟﾘｽ食品組成物	１６．３	同　上	同　上	同　上
実施例５のﾌﾟﾛﾎﾟﾘｽ食品組成物	１９．２	同　上	同　上	同　上
実施例６のﾌﾟﾛﾎﾟﾘｽ食品組成物	１６．１	同　上	同　上	同　上
実施例７のﾌﾟﾛﾎﾟﾘｽ食品組成物	１８．８	同　上	同　上	同　上
実施例８のﾌﾟﾛﾎﾟﾘｽ食品組成物	１８．９	同　上	同　上	同　上
実施例９のﾌﾟﾛﾎﾟﾘｽ食品組成物	１７．５	同　上	同　上	同　上
実施例１０のﾌﾟﾛﾎﾟﾘｽ食品組成物	１６．２	同　上	同　上	同　上

【００３９】表５から明らかなように実施例３～１０のプロポリス食品組成物は、いずれも、プロポリス成分の抽出効率が良好であり、また目的とする水親和性も十分であることが分かる。

【００４０】応用例１

撹拌機および温度計を付したビーカーに、水１２重量部、グリセリン６４．７重量部、ステアリン酸８重量部および水酸化ナトリウム０．３重量部を仕込み、徐々に加熱しながら混合して、８０℃で乳液を調製した。次いで、内温を５０℃まで下げたのち、実施例１で得られた固形状残渣１５重量部を投入し、１０００ｒｐｍで３０分間均一に混合させることにより、ディスパージョン型の汚れ落としクリームを作製した。

【００４１】比較応用例１

応用例１において、実施例１で得られた固形状残渣の代わりに、比較例１で得られた固形状残渣を用いた以外は、応用例１と同様にして、ディスパージョン型の汚れ落としクリームを作製した。

【００４２】応用例１および比較応用例１の汚れ落としクリームを、それぞれ１０ｇずつガラス製シリンダーに移し取り、２５℃に調整した恒温槽に１か月静置させて、ディスパージョンの安定性を観察した。

【００４３】一方、製造直後のものについて、１日実働８時間、自動車修理工場で現場作業している１８～６０才の男性の中から、無作為に選んだ２０人を、各汚れ落としクリームについて１０人ずつに分けて、作業終了後に腕と手に使用してもらい、次いで、布タオルで拭き取ってもらった後の汚れ落とし効率、肌触わりおよび使用後の感覚を聞かせてもらった。これらの結果を表６に示す。

【００４４】

【表６】

表6

試　料	ﾃﾞｨｽﾊﾟｰｼﾞｮﾝ安定性（２５℃、１ヶ月後）	汚れ落とし効果　1)	肌触わり　2)	使用後の感覚　3)
応用例１の汚れ落としクリーム	均一状態	○	○	○
比較応用例１の汚れ落としクリーム	底部に沈殿物形成	△	×	△

【００４５】（注）

1）汚れ落とし効果の評価基準

○：１０人の試験者中、８人以上の者が良好と認めたもの

△：１０人の試験者中、５～７人が良好と認めたもの

×：１０人の試験者中、良好と認めた者が４人以下のも

の
２）肌触わりの評価基準
○：１０人の試験者中、８人以上の者が良好と認めたもの
△：１０人の試験者中、５～７人の者が良好と認めたもの
×：１０人の試験者中、６人以上の者が肌触わり不良と認めたもの
３）使用後の感覚の評価
○：１０人の試験者中、８人以上の者がしっとりすると認めたもの
△：１０人の試験者中、５～７人がしっとりすると認めたもの
×：１０人の試験者中、しっとりすると認めた者が４人以下のもの
【００４６】応用例２
海抜３００ｍの関東ローム層畑地１２m^2の表層２０ｃｍまでの所に、実施例２で得られた固形状残渣２ｋｇを平均的に散布したのち、ラベンダーの苗を５０本植えて、夏期３ケ月間生育させた。
【００４７】比較応用例２
応用例２における場所に隣接する関東ローム層畑地１２m^2の表層２０ｃｍまでの所に、比較例２で得られた固形状残渣２ｋｇを平均的に散布したのち、ラベンダーの苗を５０本植えて、応用例２と同一時期３ケ月間生育させた。また、同時期に、応用例２および比較応用例２に隣接する関東ローム層畑地１２m^2にラベンダーの苗を５０本植えて、そのまま３ケ月間生育させた。それぞれの生育状況をまとめて、表７に示す。
【００４８】
【表７】

表7

試　料	頭頂の花部までの長さ			茎部の状態 1)
	45cmより高いもの	35～45cmのもの	35cm未満のもの	
無処理地区のラベンダー	０本／５０本	１２本／５０本	３８本／５０本	C
応用例２のラベンダー	２３本／５０本	２６本／５０本	１本／５０本	A
比較応用例２のラベンダー	３本／５０本	２８本／５０本	１９本／５０本	B

【００４９】（注）
１）茎部の状態の評価基準
Ａ：５０本中、４０本以上が頑丈に上部に向けて伸びているもの
Ｂ：５０本中、３０～３９本が頑丈に上部に向けて伸びているもの
Ｃ：頑丈に上部に向けて伸びているものが２９本以下のもの
表７から明らかなように、応用例２におけるラベンダーの生育状況が良好であることが分かる。
【００５０】
【発明の効果】本発明によれば、予め出発原料のプロポリス原塊の表面にポリオール・脂肪酸エステル系乳化剤の水系ミセル溶液を吸着させておいてから、エチルアルコールで抽出処理することにより、ミセル触媒機構とレビンダー効果が働いて、プロポリス原塊の破砕を行なうと同時に、包み込まれていた成分を迅速かつ効率良く取り出し、さらに、系内での凝集並びに再付着を抑制する。
【００５１】また、本発明の製造方法における中間工程時のミセル溶液吸着プロポリス原塊並びに最終工程において分離して得た抽出残渣も浸透力を保持する。したがって、本発明を実施することにより、アルコール抽出製品の抽出効率が上がり、しかも食しやすい水親和性へと状態変化がなされるだけでなく、従来の単純アルコール抽出プロポリス食品の製造において産業廃棄物になっていたプロポリス原塊中の不溶性物質の活用の可能性も見出されるので、大きな利点となる。
